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PCT/GB00/03281

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF THE RECORDING OF A CHANGE

(PCT Rule 92bis.1 and Administrative Instructions, Section 422)

To:

LEAMAN, Keith
QED I.P. Services Limited
Dawley Road
Hayes
Middlesex UB3 1HH
ROYAUME-UNI

Date of mailing (day/month/year)
07 June 2001 (07.06.01)

Applicant's or agent's file reference
12835/WO

IMPORTANT NOTIFICATION

International application No.
PCT/GB00/03281

International filing date (day/month/year)
24 August 2000 (24.08.00)

1. The following indications appeared on record concerning:

☐ the applicant ☐ the inventor ☒ the agent ☐ the common representative

Name and Address
WALKER, Neville, Daniel, Alan
QED I.P. Services Limited
Dawley Road
Hayes
Middlesex UB3 1HH
United Kingdom

State of Nationality

State of Residence

Telephone No.
00 44 20 8848 6692

Facsimile No.
00 44 20 8848 6469

Teleprinter No.

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☒ the person ☐ the name ☐ the address ☐ the nationality ☐ the residence

Name and Address
LEAMAN, Keith
QED I.P. Services Limited
Dawley Road
Hayes
Middlesex UB3 1HH
United Kingdom

State of Nationality

State of Residence

Telephone No.
00 44 20 8848 6633

Facsimile No.
00 44 20 8848 6469

Teleprinter No.

3. Further observations, if necessary:

4. A copy of this notification has been sent to:

☒ the receiving Office
☐ the International Searching Authority
☒ the International Preliminary Examining Authority
☐ the designated Offices concerned
☒ the elected Offices concerned
☐ other:

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer
Dominique DELMAS

Telephone No.: (41-22) 338.83.38

Form PCT/IB/306 (March 1994)

004074716

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

Commissioner
US Department of Commerce
United States Patent and Trademark
Office, PCT
2011 South Clark Place Room
CP2/5C24
Arlington, VA 22202
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 07 June 2001 (07.06.01)	
International application No. PCT/GB00/03281	**Applicant's or agent's file reference** 12835/WO
International filing date (day/month/year) 24 August 2000 (24.08.00)	**Priority date** (day/month/year) 24 August 1999 (24.08.99)
Applicant DODGSON, John, Robert et al	

1. The designated Office is hereby notified of its election made:

[X] in the demand filed with the International Preliminary Examining Authority on:

20 March 2001 (20.03.01)

[ ] in a notice effecting later election filed with the International Bureau on:

2. The election [X] was

[ ] was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO **34, chemin des Colombettes** **1211 Geneva 20, Switzerland**	Authorized officer Dominique DELMAS
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	**FOR FURTHER ACTION** see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
12835/WO		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/GB 00/03281	24/08/2000	24/08/1999
Applicant		
CENTRAL RESEARCH LABORATORIES LTD.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language,** the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

[ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

[ ] contained in the international application in written form.

[ ] filed together with the international application in computer readable form.

[ ] furnished subsequently to this Authority in written form.

[ ] furnished subsequently to this Authority in computer readble form.

[ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

[ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).

3. [ ] **Unity of invention is lacking** (see Box II).

4. With regard to the **title,**

[X] the text is approved as submitted by the applicant.

[ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract,**

[X] the text is approved as submitted by the applicant.

[ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. 3

[X] as suggested by the applicant.

[ ] None of the figures.

[ ] because the applicant failed to suggest a figure.

[ ] because this figure better characterizes the invention.

INTERNATIONAL SEARCH REPORT

International Application No
GB 00/03281

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 G01N27/49

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 G01N H01R

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 914 019 A (DODGSON JOHN ROBERT ET AL) 22 June 1999 (1999-06-22) cited in the application column 5, line 27 - line 30; figure 1	1,18,30, 31
A	DE 43 04 747 A (SHINETSU POLYMER CO) 9 September 1993 (1993-09-09) abstract; figure 2	1,31
A	GB 2 299 863 A (DRAEGERWERK AG) 16 October 1996 (1996-10-16) abstract; figure 1	1
A	FR 2 499 246 A (COAL INDUSTRY PATENTS LTD) 6 August 1982 (1982-08-06) page 6, line 26 -page 7, line 38; figure 1	1

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
13 February 2001	20/02/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Duchatellier, M

1

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International Application No
[redacted]/GB 00/03281

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	WO 98 25139 A (DODGSON JOHN ROBERT ;ROBINS IAN (GB); AUSTEN MALCOLM TRAYTON (GB);) 11 June 1998 (1998-06-11) abstract	23

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
[redacted]/GB 00/03281

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 5914019	A	22-06-1999	AU	3812695 A	31-05-1996
			CA	2204413 A	17-05-1996
			CN	1162357 A,B	15-10-1997
			DE	69514943 D	09-03-2000
			DE	69514943 T	05-10-2000
			EP	0871874 A	21-10-1998
			ES	2144148 T	01-06-2000
			FI	971846 A	02-06-1997
			WO	9614576 A	17-05-1996
			JP	10508696 T	25-08-1998
			DK	871874 T	26-06-2000
DE 4304747	A	09-09-1993	JP	6318478 A	15-11-1994
			JP	7013901 B	15-02-1995
			JP	1961812 C	25-08-1995
			JP	6076877 A	18-03-1994
			JP	6085336 B	26-10-1994
			JP	2502900 B	29-05-1996
			JP	7022098 A	24-01-1995
			GB	2265500 A,B	29-09-1993
			KR	9704764 B	03-04-1997
			US	5371327 A	06-12-1994
GB 2299863	A	16-10-1996	DE	19514214 A	24-10-1996
			FR	2733052 A	18-10-1996
			US	5702576 A	30-12-1997
FR 2499246	A	06-08-1982	DE	3203362 A	30-09-1982
			GB	2094005 A,B	08-09-1982
			JP	2077918 C	09-08-1996
			JP	6056376 B	27-07-1994
			JP	57147048 A	10-09-1982
			US	4406770 A	27-09-1983
WO 9825139	A	11-06-1998	CN	1244258 A	09-02-2000
			EP	0943092 A	22-09-1999

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
PQ12835		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/03281	24/08/2000	24/08/1999
International Patent Classification (IPC) or national classification and IPC G01N27/49		
Applicant CENTRAL RESEARCH LABORATORIES LTD.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 10 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☒ Certain defects in the international application
   - VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
20/03/2001	26.10.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Roetsch, P Telephone No. +49 89 2399 2548

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03281

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:
Description, pages:

1-14	as originally filed

Claims, No.:

1-31	as originally filed

Drawings, sheets:

1/2-2/2	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   - ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
   - ☐ the language of publication of the international application (under Rule 48.3(b)).
   - ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   - ☐ contained in the international application in written form.
   - ☐ filed together with the international application in computer readable form.
   - ☐ furnished subsequently to this Authority in written form.
   - ☐ furnished subsequently to this Authority in computer readable form.
   - ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
   - ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   - ☐ the description, pages:
   - ☐ the claims, Nos.:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT International application No. PCT/GB00/03281

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☐ the entire international application.

☒ claims Nos. 1-30.

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 1-30 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☐ no international search report has been established for the said claims Nos. .

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N) Yes: Claims 31

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03281

	No:	Claims	
Inventive step (IS)	Yes:	Claims	31
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	31
	No:	Claims	

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

INTERNATIONAL PRELIMINARY EXAMINATION REPORT - SEPARATE SHEET International application No. PCT/GB00/03281

Reference is made to the following documents:

D1: US-A-5 914 019 (DODGSON JOHN ROBERT ET AL) 22 June 1999 (1999-06-22) cited in the application

D2: US-A-5 314 605 (MATTHIESSEN HANS) 25 November 1992 (1992-11-25) cited in the application *

* **D2** is not cited in the International Search Report but is cited in the description of the present application.

Ad Section III

The questions wether the subject-matter of claims 1-30 appears to be novel, to involve an inventive step, or to be industrially applicable have not been examined because these claims and in particular the independent claims 1, 18 and 30 are so unclear (see section VIII below) that no meaningful opinion could be formed.

Ad Section V

1) The invention pertains to a method of forming an electrical pathway across a microporous membrane (independent claim 31).

2) **NOVELTY (independent claim 31)**

2.1) Document **D2** (cited by the Applicant), which is considered to represent the closest prior art, discloses (cf. figure 1) a gas sensor assembly with a ceramic porous membrane (2) through which holes have been formed and electrodes (14) passed trough said holes for forming an electrical pathway across the membrane.

2.2) Following features from claim 31 are not disclosed in document **D2**: the electrical pathway through the gas porous membrane is formed by maintaining sufficient heat to melt a conductive material; urging the melted conductive material through pores of the membrane at a first surface by establishing a pressure differential across the surfaces; controlling the heat and pressure differential until the conductive material emerges at the second surface; and allowing the material to cool so as to form a continuous, electrically conductive pathway from the first to

the second surface whilst preserving the liquid impermeability and gas permeability characteristics of the membrane.

2.3) Claim 31 thus meets the requirements of Article 33(2) PCT.

3) **INVENTIVE STEP (independent claim 31)**

3.1) When starting from **D2**, the problem can be seen as how to provide an electrical pathway across a porous membrane that is less prone to leaking.

3.2) The present invention solves the problem by urging melted conductive material through pores of a membrane in order to form an electrical pathway across the membrane, whilst preserving the liquid impermeability of the membrane.

3.3) Neither document **D2** nor one of the remaining documents from the International Search Report discloses a method of forming an electrical pathway across a membrane by urging a melted conductive material through pores of said membrane.

3.4) Claim 31 thus meet the requirements of Article 33(3) PCT.

Ad Section VII

1) The description on page 10, lines 10-29 is unclear since it refers to figure 3 (page 10, line 10) but uses wrong reference numbers.

2) The term "membrane" should have been used throughout the complete description instead of "substrate" (cf. also paragraphs VIII.4. and VIII.16. below).

3) The independent claims are not in the two-part form in accordance with Rule 6.3(b) PCT, which in the present case would be appropriate, with those features known in combination from the prior art (document **D1** or **D2**) being placed in the preamble (Rule 6.3(b)(i) PCT) and with the remaining features being included in the characterising part (Rule 6.3(b)(ii) PCT).

4) The features of the independent claims are not provided with reference signs placed in parentheses (Rule 6.2(b) PCT).

Ad Section VIII

1) Independent **claim 1** discloses a method of manufacturing a gas sensor, but in fact merely defines a method of providing an electrical pathway between only one electrode and an electrical contact. The other essential features concerning at least the second electrode (counter electrode in the case of an electrochemical sensor) and the contact between the electrolyte and both electrodes are missing in claim 1.
Since independent claim 1 does not contain these features it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

2) In order to harmonize the claims, the expression "external electrical contact" (cf. claims 8 and 18) should have been used instead of "electrical contact" or "external connection means" in **claims 1, 8, 11-13, 18-19** and **25-27**.

3) **Claims 2-5** and **11-13** refer back to claim 1, where only one electrode has been defined. The term "the electrodes" in those claims is thus unclear.

4) The term "the substrate" in **claims 6-17** has no proper antecedent.

5) In order to harmonize the claims, the expression "impregnated" (cf. claim 1) should have been used instead of "introduced" in **claims 6-10**.

6) **Claim 6** refers back to claim 1, where "the wicking means" were not defined. The referring back thus is unclear.

7) **Claims 7** and **16** leave a doubt whether "an electrode" is the electrode defined in claim 1 or an additional electrode.

8) The feature in **claim 9** "the conductive material is introduced into the substrate via

the substrate" is contradictory in itself and as such renders the claim unclear.

9) **Claim 10** leaves a doubt whether "conductive material" is the conductive material defined in claim 1 or an additional conductive material.

10) The skilled person would not know what is meant by the feature of **claim 11** "filtering in selected areas from a suspension placed onto the substrate".

11) The expression "the electrodes are formed on the opposite faces of the substrate to the external connection means" in **claim 12** is incomprehensible.

12) **Claim 16** does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved which merely amounts to a statement of the underlying problem: "the permeability of at least one region of the substrate to gas is decreased in order to limit the amount of gas reaching an electrode". The technical features necessary for achieving this result should be added.

13) It is clear from the description on page 2, lines 19-21 that the following features are essential to the definition of the invention:
(1) The membrane seals the reservoir (cf. also claim 1).
(2) The membrane is a gas porous membrane (see also paragraph 15 below)
Since independent **claim 18** does not contain these features it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

14) **Claims 18** leaves a doubt whether "an electrode" and "the electrode" (page 17, lines 7,8 and 11) correspond to the "first and second electrode" defined before (page 17, line 3) or to an additional electrode.
The same deficiency arises for the term "the material" (page 17, line 10) and "the conductive material" (page 17, line 9).

15) As explained below, some of the features in the apparatus **claims 18, 21** and **28**

relate to a method of using the apparatus rather than clearly defining the apparatus in terms of its technical features. The intended limitations are therefore not clear from these claims, contrary to the requirements of Article 6 PCT.

- **Claim 18**: "a reservoir which, in use, contains liquid electrolyte for contacting the first and second electrodes".
- **Claim 21**: "a counter electrode which performs an electrochemical reaction with oxygen".
- **Claim 28**: "the wicking means ... which in use, it contacts both the electrolyte and the electrodes"

16) In order to harmonize the claims, the expression "membrane" (cf. claim 1) should have been used instead of "substrate" in **claims 18** and **27** (cf. also paragraph VII.2. above).

17) **Claim 18** leaves a doubt how the "electrical contact" is arranged relative to the membrane in the gas sensor assembly.

18) In **claim 18**, the feature "a portion of the electrode and a portion of the substrate substantially adjacent thereto, is impregnated with the conductive material" contradicts the previous feature "conductive material disposed between an electrode and the external electrical contact":

19) The expression "the desired electrochemical reaction" in **claim 20** has no proper antecedent.

20) **Claims 20** and **28** do not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not defined. The claims attempt to define the gas sensor by further unclaimed features namely the electrolyte (see the Guidelines PCT-Section IV-III, 4.8a).

21) The term "conductive mass" in **claims 24-25** has no proper antecedent. It seems that "conductive material" (cf. claim 18) was meant.

22) The feature of **claim 25**, that "the conductive mass is a plug, pin, or other shaped component suitable", is not referred to in the description. The description only

states that "the conductive material may be in the form of a plug, pin or other shaped material" ; since the conductive material is impregnated in the membrane (cf. claim 18) it is not part of the invention to insert an electrically conductive pin, plug or other shaped material in the membrane. Claim **25** is therefore not supported by the description as required by Article 6 PCT.

23) **Claim 25** leaves the reader in doubt whether all the electrodes defined before are connected with the conductive mass to the external connection means or whether only one electrode (cf. claim 18, page 17, line 7) is connected to said external connection means.

24) **Claim 30** contains a reference to the drawings. According to Rule 6.2(a) PCT, claims should not contain such references except where absolutely necessary, which is not the case here. **Claim 30** should have been deleted.

25) **Claim 31** leaves a doubt whether "a first surface" and "the second surface" are the "first and second major surfaces" as defined before or additional surfaces.

26) **Claim 31** does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved which merely amounts to a statement of the underlying problem: "whilst preserving the liquid impermeability and gas permeability characteristics of the membrane". The technical features necessary for achieving this result should have been added.

WO 01/14868
PCT/GB00/03281

PATENT COOPERATION TREATY

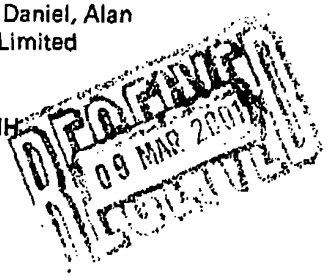
From the INTERNATIONAL BUREAU

PCT

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

To:
WALKER, Neville, Daniel, Alan
QED I.P. Services Limited
Dawley Road
Hayes
Middlesex UB3 1HH
ROYAUME-UNI



Date of mailing (day/month/year)
01 March 2001 (01.03.01)

Applicant's or agent's file reference	IMPORTANT NOTICE
12835/WO	

International application No.	International filing date (day/month/year)	Priority date (day/month/year)
PCT/GB00/03281	24 August 2000 (24.08.00)	24 August 1999 (24.08.99)

Applicant
CENTRAL RESEARCH LABORATORIES LTD. et al

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
   AU,KP,KR,US

   In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
   AE,AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CN,CR,CU,CZ,DE,DK,DM,EA,EE,EP,ES,FI,GB,GD,GE,GH,GM,HR,HU,ID,IL,IN,IS,JP,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MA,MD,MG,MK,MN,MW,MX,NO,NZ,OA,PL,PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,TZ,UA,UG,UZ,VN,YU,ZA,ZW
   The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
   01 March 2001 (01.03.01) under No. WO 01/14868

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38

PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

14

Applicant's or agent's file reference PQ12835	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/GB00/03281	International filing date *(day/month/year)* 24/08/2000	Priority date *(day/month/year)* 24/08/1999
International Patent Classification (IPC) or national classification and IPC G01N27/49		
Applicant CENTRAL RESEARCH LABORATORIES LTD.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 10 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☒ Certain defects in the international application
   - VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
20/03/2001	26.10.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Roetsch, P Telephone No. +49 89 2399 2548

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03281

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:
Description, pages:

1-14 as originally filed

Claims, No.:

1-31 as originally filed

Drawings, sheets:

1/2-2/2 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☐ the entire international application.

☒ claims Nos. 1-30.

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 1-30 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☐ no international search report has been established for the said claims Nos. .

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N) Yes: Claims 31

	No:	Claims	
Inventive step (IS)	Yes:	Claims	31
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	31
	No:	Claims	

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Reference is made to the following documents:

D1: US-A-5 914 019 (DODGSON JOHN ROBERT ET AL) 22 June 1999 (1999-06-22) cited in the application

D2: US-A-5 314 605 (MATTHIESSEN HANS) 25 November 1992 (1992-11-25) cited in the application *

* **D2** is not cited in the International Search Report but is cited in the description of the present application.

Ad Section III

The questions wether the subject-matter of claims 1-30 appears to be novel, to involve an inventive step, or to be industrially applicable have not been examined because these claims and in particular the independent claims 1, 18 and 30 are so unclear (see section VIII below) that no meaningful opinion could be formed.

Ad Section V

1) The invention pertains to a method of forming an electrical pathway across a microporous membrane (independent claim 31).

2) **NOVELTY (independent claim 31)**

2.1) Document **D2** (cited by the Applicant), which is considered to represent the closest prior art, discloses (cf. figure 1) a gas sensor assembly with a ceramic porous membrane (2) through which holes have been formed and electrodes (14) passed trough said holes for forming an electrical pathway across the membrane.

2.2) Following features from claim 31 are not disclosed in document **D2**: the electrical pathway through the gas porous membrane is formed by maintaining sufficient heat to melt a conductive material; urging the melted conductive material through pores of the membrane at a first surface by establishing a pressure differential across the surfaces; controlling the heat and pressure differential until the conductive material emerges at the second surface; and allowing the material to cool so as to form a continuous, electrically conductive pathway from the first to

the second surface whilst preserving the liquid impermeability and gas permeability characteristics of the membrane.

2.3) Claim 31 thus meets the requirements of Article 33(2) PCT.

3) **INVENTIVE STEP (independent claim 31)**

3.1) When starting from **D2,** the problem can be seen as how to provide an electrical pathway across a porous membrane that is less prone to leaking.

3.2) The present invention solves the problem by urging melted conductive material through pores of a membrane in order to form an electrical pathway across the membrane, whilst preserving the liquid impermeability of the membrane.

3.3) Neither document **D2** nor one of the remaining documents from the International Search Report discloses a method of forming an electrical pathway across a membrane by urging a melted conductive material through pores of said membrane.

3.4) Claim 31 thus meet the requirements of Article 33(3) PCT.

Ad Section VII

1) The description on page 10, lines 10-29 is unclear since it refers to figure 3 (page 10, line 10) but uses wrong reference numbers.

2) The term "membrane" should have been used throughout the complete description instead of "substrate" (cf. also paragraphs VIII.4. and VIII.16. below).

3) The independent claims are not in the two-part form in accordance with Rule 6.3(b) PCT, which in the present case would be appropriate, with those features known in combination from the prior art (document **D1** or **D2**) being placed in the preamble (Rule 6.3(b)(i) PCT) and with the remaining features being included in the characterising part (Rule 6.3(b)(ii) PCT).

4) The features of the independent claims are not provided with reference signs placed in parentheses (Rule 6.2(b) PCT).

Ad Section VIII

1) Independent **claim 1** discloses a method of manufacturing a gas sensor, but in fact merely defines a method of providing an electrical pathway between only one electrode and an electrical contact. The other essential features concerning at least the second electrode (counter electrode in the case of an electrochemical sensor) and the contact between the electrolyte and both electrodes are missing in claim 1.
Since independent claim 1 does not contain these features it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

2) In order to harmonize the claims, the expression "external electrical contact" (cf. claims 8 and 18) should have been used instead of "electrical contact" or "external connection means" in **claims 1, 8, 11-13, 18-19** and **25-27**.

3) **Claims 2-5** and **11-13** refer back to claim 1, where only one electrode has been defined. The term "the electrodes" in those claims is thus unclear.

4) The term "the substrate" in **claims 6-17** has no proper antecedent.

5) In order to harmonize the claims, the expression "impregnated" (cf. claim 1) should have been used instead of "introduced" in **claims 6-10**.

6) **Claim 6** refers back to claim 1, where "the wicking means" were not defined. The referring back thus is unclear.

7) **Claims 7** and **16** leave a doubt whether "an electrode" is the electrode defined in claim 1 or an additional electrode.

8) The feature in **claim 9** "the conductive material is introduced into the substrate via

the substrate" is contradictory in itself and as such renders the claim unclear.

9) **Claim 10** leaves a doubt whether "conductive material" is the conductive material defined in claim 1 or an additional conductive material.

10) The skilled person would not know what is meant by the feature of **claim 11** "filtering in selected areas from a suspension placed onto the substrate".

11) The expression "the electrodes are formed on the opposite faces of the substrate to the external connection means" in **claim 12** is incomprehensible.

12) **Claim 16** does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved which merely amounts to a statement of the underlying problem: "the permeability of at least one region of the substrate to gas is decreased in order to limit the amount of gas reaching an electrode". The technical features necessary for achieving this result should be added.

13) It is clear from the description on page 2, lines 19-21 that the following features are essential to the definition of the invention:
(1) The membrane seals the reservoir (cf. also claim 1).
(2) The membrane is a gas porous membrane (see also paragraph 15 below)
Since independent **claim 18** does not contain these features it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

14) **Claims 18** leaves a doubt whether "an electrode" and "the electrode" (page 17, lines 7,8 and 11) correspond to the "first and second electrode" defined before (page 17, line 3) or to an additional electrode.
The same deficiency arises for the term "the material" (page 17, line 10) and "the conductive material" (page 17, line 9).

15) As explained below, some of the features in the apparatus **claims 18, 21** and **28**

relate to a method of using the apparatus rather than clearly defining the apparatus in terms of its technical features. The intended limitations are therefore not clear from these claims, contrary to the requirements of Article 6 PCT.

- **Claim 18**: "a reservoir which, in use, contains liquid electrolyte for contacting the first and second electrodes".
- **Claim 21**: "a counter electrode which performs an electrochemical reaction with oxygen".
- **Claim 28**: "the wicking means ... which in use, it contacts both the electrolyte and the electrodes"

16) In order to harmonize the claims, the expression "membrane" (cf. claim 1) should have been used instead of "substrate" in **claims 18** and **27** (cf. also paragraph VII.2. above).

17) **Claim 18** leaves a doubt how the "electrical contact" is arranged relative to the membrane in the gas sensor assembly.

18) In **claim 18**, the feature "a portion of the electrode and a portion of the substrate substantially adjacent thereto, is impregnated with the conductive material" contradicts the previous feature "conductive material disposed between an electrode and the external electrical contact":

19) The expression "the desired electrochemical reaction" in **claim 20** has no proper antecedent.

20) **Claims 20** and **28** do not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not defined. The claims attempt to define the gas sensor by further unclaimed features namely the electrolyte (see the Guidelines PCT-Section IV-III, 4.8a).

21) The term "conductive mass" in **claims 24-25** has no proper antecedent. It seems that "conductive material" (cf. claim 18) was meant.

22) The feature of **claim 25**, that "the conductive mass is a plug, pin, or other shaped component suitable", is not referred to in the description. The description only

states that "the conductive material may be in the form of a plug, pin or other shaped material" ; since the conductive material is impregnated in the membrane (cf. claim 18) it is not part of the invention to insert an electrically conductive pin, plug or other shaped material in the membrane. Claim **25** is therefore not supported by the description as required by Article 6 PCT.

23) **Claim 25** leaves the reader in doubt whether all the electrodes defined before are connected with the conductive mass to the external connection means or whether only one electrode (cf. claim 18, page 17, line 7) is connected to said external connection means.

24) **Claim 30** contains a reference to the drawings. According to Rule 6.2(a) PCT, claims should not contain such references except where absolutely necessary, which is not the case here. **Claim 30** should have been deleted.

25) **Claim 31** leaves a doubt whether "a first surface" and "the second surface" are the "first and second major surfaces" as defined before or additional surfaces.

26) **Claim 31** does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved which merely amounts to a statement of the underlying problem: "whilst preserving the liquid impermeability and gas permeability characteristics of the membrane". The technical features necessary for achieving this result should have been added.

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Published:
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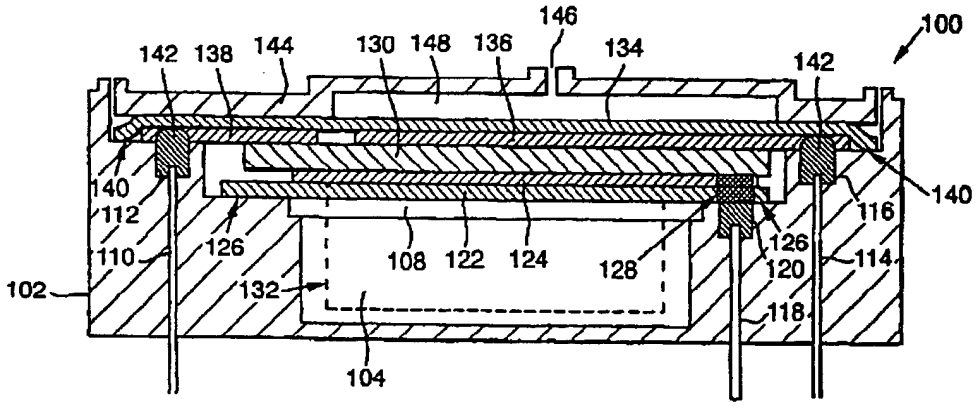
For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: GAS SENSOR AND ITS METHOD OF MANUFACTURE



(57) Abstract: The invention relates to a gas sensor and its method of manufacture. Electrochemical gas sensors usually comprise an external housing, which acts as a reservoir for electrolyte; a wick to keep electrodes wetted with the electrolyte and external electrical terminals, for making electrical contact with the electrodes. Typically a gas permeable/microporous membrane has been used to seal a gas sensor in order to prevent leakage of electrolyte. A problem with existing sensors has been that there was a risk of electrolyte leaking through the membrane around the region where electrical connector passed therethrough. The present invention overcomes this by providing a method of urging conductive polymer through the membrane under controlled conditions of heat and pressure, thereby ensuring the integrity of the membrane remains intact whilst defining an electrically/conductive pathway therethrough.

WO 01/14868 A2

GAS SENSOR AND ITS METHOD OF MANUFACTURE

The present invention relates to a gas sensor, and to its method of manufacture. It relates particularly, but not exclusively, to an electrochemical gas sensor for sensing carbon monoxide (CO) gas.

An electrochemical gas sensor for sensing an oxidisible or reducible gas, such as carbon monoxide, usually includes a sensing electrode, a counter electrode and a diffusion barrier. The diffusion barrier allows gas to be sensed, to pass to the sensing electrode. In one type of gas sensor, as described, for example, in the Applicant's copending International Patent Application No. WO-A1-9614576, the sensing and counter electrodes are located on a gas permeable membrane and are in contact with an electrolyte.

In terms of physical construction, electrochemical gas sensors usually comprise an external housing, which acts as a reservoir for electrolyte; a wick, to keep the electrolyte in contact with the electrodes; and external electrical terminals, which make electrical contact with the electrodes.

During operation of the aforementioned gas sensor, an electrochemical reaction occurs at the sensing electrode with the gas to be sensed, and a reaction also occurs with oxygen at the counter electrode. Electric current is carried through the electrolyte by ions produced in these reactions, and the amount of current indicates the concentration of the gas being sensed. A further electrode (the reference electrode) may be employed, for example, in combination with a potentiostat circuit, to maintain a constant potential difference between the sensing electrode and the electrolyte. This increases the stability of operation of the gas sensor. Electrodes are connected to external current sensors via electrical terminals.

External electrical terminals are usually formed from brass or copper pins. Brass and copper both react with the acid electrolyte, and so the gas sensor has to be specially designed so that the pins do not come into contact with the electrolyte. Platinum does not react with acid, and so platinum strips can be used to form an electrical path between the electrodes and external sensors and/or an external electricity supply. However, platinum strips are commonly placed in a seal region between the housing and the gas permeable membrane, and electrolyte can leak from this region. Platinum is also expensive, and so gas sensors having platinum terminals are expensive to manufacture.

Another example of a gas sensor is described in US Patent US-A-5314605 (Dragerwerk). The aforementioned US Patent describes a gas permeable region through which holes have been formed. Electrodes pass through the holes. No matter how carefully the region between the periphery of each hole and the electrode is sealed, there is a risk of electrolyte leaking through this seal.

An aim of the present invention is to provide a gas sensor that is cheaper to manufacture than existing gas sensors. Another aim of the invention is to provide a gas sensor that is less prone to leaking than existing gas sensors.

According to a first aspect of the present invention there is provided a method of manufacturing a gas sensor having a housing containing a reservoir which in use receives an electrolyte, the method comprising the steps of: impregnating a gas porous membrane with a conductive material, so that said conductive material defines an electrical pathway between an electrical contact on a first surface of the membrane and an electrode on a second surface of the membrane and arranging the gas porous membrane to seal the reservoir.

Thus in accordance with a first aspect of the invention, a simple and reliable means is provided for connecting one or more electrodes, located within a sealed reservoir of the gas sensor, to an electrical pathway outside the sensor: the method avoiding the use of expensive platinum terminals and one which produces a gas sensor which is less prone to leaking.

Preferably the conductive material comprises a conductive polymer and is introduced into the pores of a microporous membrane under conditions of heat and pressure.

The method may also include the step of attaching a wicking means to one or more of the electrodes. The wicking means ensures that each electrode is/are kept in contact with the electrolyte irrespective of the orientation of the sensor once installed. The wicking means may be pressed or sintered to the or each electrode at a temperature of between 300°C and 370°C, most preferably between 320°C and 370°C. The exact temperature depends on the nature of the wicking means, the electrode material, and the substrate. Attachment of a wicking means may be performed before any melted conductive polymer is introduced, in which case the wicking means may have at least one aperture therein through which melted polymer can pass to an electrode.

According to a second aspect of the invention there is provided a method of forming an electrical pathway across a microporous membrane having first and second major surfaces; which membrane in use is impervious to liquid and permeable to gas, comprising the steps of: maintaining sufficient heat to melt a conductive material; urging the melted conductive material through pores of the membrane at a first surface by establishing a pressure differential across the surfaces; controlling the heat and pressure differential until the conductive material emerges at the second surface; and allowing the material to cool so as to form a continuous, electrically conductive pathway from the first to the second surface whilst preserving the liquid impermeability and gas permeability characteristics of the membrane.

Preferably the microporous membrane thereby formed is incorporated into an electrochemical cell which may be incorporated into a gas sensor.

Conductive material may be introduced into the substrate via the wicking means, via the electrodes, via the substrate, or via a combination of these.

The conductive material preferably includes conductive polymer. On cooling and solidification of the conductive material, an electrical path is formed between the electrode and the electrical contact or external connection means. Electric current generated in use, at the sensing electrode, may thus pass via the microporous membrane, by way of the conductive polymer to the external connection means, and then to a suitable electronic device (or current source in the case of a test gas generator) where the amount of current generated at the sensing electrode can be measured.

The first and second electrodes are preferably formed from a porous electrically conductive material containing PTFE or similar polymeric binder. Electrodes may also contain particles of catalyst, and optional, additional catalyst support material and material to enhance conductivity.

Electrodes may be formed on the substrate by, for example, screen printing, filtering in selected areas from a suspension placed onto the substrate, by spray coating, ink jet printing, sintering, or any other method suitable for producing a patterned deposition of solid material. Deposition might be of a single material, or of more than one material sequentially in layers so as, for example, to vary the properties of the electrode material through its thickness.

Preferably first and second electrodes are formed on an opposite surface of the substrate to the external electrical contact means. Alternatively, the first and second electrodes and the external electrical contact means, may be formed on the same side of the substrate.

The substrate may be bonded to the housing using adhesive. Alternatively, a mechanical means such as a snap-link may be used. It is preferred, however, to

employ heat and/or pressure to bond the substrate to the housing. The housing preferably comprises a synthetic plastics material with a lower melting point than the substrate. When the substrate and the housing are fixed together using heat and/or pressure, housing material impregnates the substrate thereby forming a strong mechanical bond which is also impervious to the electrolyte.

A cap member having a diffusion barrier may also be provided. The substrate is positioned between the cap member and the housing. Heat and/or pressure (or other suitable method) is then applied to seal the sensor assembly. If a cap member is not used, then the permeability of at least one region of the substrate may be modified in order to control the amount of gas reaching the electrodes. This may be achieved by use of a material with the required porosity, or the porosity may be decreased either by i) compressing the region, or ii) by impregnating the region(s) with, for example, wax, polymer, or a wax/polymer mix.

According to a further aspect of the invention there is provided a gas sensor comprising: at least first and second electrodes formed on a planar substrate; a housing containing a reservoir which, in use, contains liquid electrolyte for contacting the first and second electrodes; an electrical contact for making external electrical connection from the gas sensor; and a conductive material disposed between an electrode and the external electrical contact, wherein at least a portion of the electrode and a portion of the substrate substantially adjacent thereto, is impregnated with the conductive material, the material forming an electrical pathway through the membrane which connects at least an electrode to the external electrical contact.

The electrodes are preferably porous planar elements. The first electrode is preferably a gas sensing (working electrode) for creating the desired electrochemical reaction between the electrolyte and the gas to be sensed. The second electrode is preferably a counter electrode which performs the counterpart electrochemical reaction with oxygen. The gas sensor may include further electrodes, such as a reference electrode and/or a test gas generating electrode.

The conductive material may be in the form of a plug, pin, or other shaped component suitable for forming an electrical path between the electrodes and an external connection means.

The external electrical contact or connection means is preferably a porous planar element which may be formed on the substrate in an identical manner to the formation of the electrodes. Alternatively, the external connection means may be formed from the same, or a similar material, to the conductive material. The external connection means may also be a metal strip, or wire, which is attached to the substrate.

The sensor may have a cap so that the substrate is disposed between the cap and the housing. In this particular arrangement, the substrate is preferably highly gas permeable and presents little or no barrier to diffusion of gas there through. In such an embodiment, diffusion of gas to the sensing electrode is preferably limited by a diffusion barrier located in the cap.

Alternatively, the sensor may have no cap, so that the substrate itself acts as a diffusion barrier and forms the upper part of the housing. In this case, porosity of the substrate in certain regions is preferably decreased in order to limit the amount of gas reaching the sensing electrode and/or the counter electrode. The substrate may be flexible, semi-rigid, or rigid.

Preferably the electrolyte is sulphuric acid or other suitable electrolyte.

Embodiments of the invention, will now be described, by way of example only, and with reference to the accompanying Figures, in which:-

Figure 1 shows a cross-section of a first gas sensor;

Figure 2 shows a cross-section of a second gas sensor; and

Figure 3 shows a sectional view through another gas sensor.

Referring to Figure 1 there is shown a sectional view of an lectrochemical gas sensor 10a in the form of a right circular cylinder, the sensor comprises a two part housing 12a and 12b, a sensing electrode 14, a counter electrode 16, and external contact tracks 28a and 28b formed on a generally circular membrane 18. Electrodes 14 and 16 are formed from a mixture of electrically conductive catalyst particles in PTFE binder, and are screen printed or filter deposited onto the surface of the membrane 18 in the form of segments, as shown in the Figure. External contacts 28a and 28b are formed by urging conductive polymer, which may be loaded with conductive non-catalytic particles, through the membrane 18.

Housing portion 12b is cylindrical with a hollow interior defining an electrolyte reservoir 20, which in use contains a liquid electrolyte 30. Electrolyte 30 is maintained in contact with the electrodes 14,16 by means of a wick 21. The electrolyte reservoir 20 is closed at the base by means of a base member 32 having a pressure relief vent closed by a porous membrane. Housing part 12a is a disc shaped cap member having an aperture 22 therein to permit atmospheric gas to diffuse to a recessed manifold area 24, and then to sensing electrode 14. The housing portions comprise a synthetic plastics material. Aperture 22 may be in the form of a diffusion barrier to control the amount of gas reaching the sensing electrode.

Membrane 18 is disc shaped and is of approximately the same diameter as lower housing portion 12b. The membrane is disposed between upper housing portion 12a and lower housing portion 12b. As the upper housing portion 12a is smaller in diameter than lower housing portion 12b, external contact tracks 28a and 28b extend beyond the edge of upper housing portion 12a, and may thus be used as an external electrical contact or connection. The external electrical contacts may be connected to a printed circuit board and a power supply by way of pins, spring clips, or wires (not shown). A solid polymer 26 is heated and forced under pressure, through the membrane so that it forms contact 28. Details of how this is achieved are described below.

Referring now to Figure 2 which shows a sectional view of the second embodiment 10b of the invention, similar parts to those of Figure 1 are denoted by the same reference numerals. In this embodiment of the invention, the upper cap member 12a is not present. The membrane 18 is of a low permeability to gases in order to define a diffusion barrier for incoming gas. Thus precise control over the rate of ingress of gas is provided. The permeability of the membrane 18 may be uniform over its entire area, or the permeability may be reduced in a particular region by, for example, pressing or impregnating certain areas of the membrane with a suitable substrate.

In gas sensor 10b, regions of the electrodes 14 and 16 and the membrane 18 are impregnated with a conductive polymer 26 such that the conductive polymer 26 protrudes through the membrane 18 to form external contacts 28a and 28b. Further external electrical contact means may then be provided.

One advantage of the gas sensor according to the present invention, over existing gas sensors is that the electrodes of sensors 10a,b do not extend between the housing and the membrane 18, which are generally the weakest part of the gas sensor assembly. Thus in gas sensors 10a and 10b, a strong seal is formed between the housing and the membrane, and electrolyte is less likely to leak from the sensor.

During operation of gas sensors 10a and 10b, gas from the environment diffuses through the membrane 18 (via aperture 22 for sensor 10a) to sensing electrode 14. If this gas contains, for example, carbon monoxide, an electrochemical reaction occurs at sensing electrode 14, and an electrochemical reaction with oxygen occurs at counter electrode 16. Current is thus carried through the electrolyte 30 by ions produced in these reactions. The size of the current indicates the concentration of carbon monoxide.

A reference electrode (not shown) may be employed in combination with a potentiostat circuit (not shown) to maintain the potential between the sensing electrode 14 and the electrolyte 30 in order to increase the stability of the sensor 10a.

The assembly of sensor 10a will now be described. Electrodes 14 and 16 are formed on the lower surface of membrane 18. External contact tracks 28a and 28b are formed on the upper surface of this membrane. The wick 21 is then sintered to the electrodes 14 and 16. Molten conductive polymer 26 is introduced into required areas of the membrane 18 via holes in the wick 21, or from the upper surface of external contract tracks 28a,b, by applying heat and pressure to force the polymer through the membrane so a contact is made between external contacts 28a and 28b and electrodes 14 and 16. On solidification of the polymer 26, an electrical path is formed between the electrolyte 30 contained with electrolyte reservoir 20 and the external contact tracks 28a and 28b.

The membrane is then positioned between upper 12a and lower housing portions 12b, and heat and pressure are applied using a press tool in order to compress the membrane and the external contacts onto the housing portions, thereby bonding the assembly together. Alternatively, one or both of the housing portions 12a,b may be bonded to the membrane 18 using adhesive.

Electrolyte is then introduced into the electrolyte reservoir 20 via aperture 32. This aperture is then plugged with an acid-tight plug (which may be gas permeable), and sealed in place using ultrasonic bonding. This ensures that electrolyte 30 does not leak from the sensor cell 10a.

The assembly of sensor 10b is similar to that of sensor 10a. Electrodes 14 and 16 are formed on the lower surface of the membrane 18. If required, the permeability to gas of regions of the membrane may be decreased, as described previously. The wick 21 is then sintered to the electrodes 14 and 16. Molten conductive polymer 26 is introduced into required areas of the membrane 18 from the upper surface of the membrane 18, by applying heat and pressure to force the material through the membrane so that, on solidification, an amount solidified conductive polymer protrudes through the membrane 18 to form external contacts across porous membrane, without altering its mechanical integrity (i.e. tearing it) but provides an

electrical pathway through membrane 28a and 28b. Further external contact means may be provided, held in place by the solidified conducting polymer.

The membrane is then positioned above lower housing portion 12b, and heat and pressure are applied using a press tool in order to compress the membrane onto the housing portion, thereby bonding the assembly together. Alternatively, the lower housing portion 12b may be bonded to the membrane 18 using adhesive. Electrolyte is then introduced into the electrolyte reservoir 20 as previously described.

Referring to Figure 3, a conductive contact or via 28 is formed by the process of impregnation of the porous membrane or substrate 18 by the conductive material in liquid form. In a preferred method, the substrate 18 is a polymeric material with open porosity, and the material to be impregnated is a polymer with lower melting point than the substrate material, loaded with conductive particles. The impregnating material 26 is forced into the pores of the substrate 18 in liquid form under pressure, so as to form a conductive mass 26 within the pores extending from one side of the substrate to the other. The mean size of the conductive particles may be smaller than that of the pores in the substrate, or may be comparable or larger, in which case the impregnation process and the substrate material are chosen to give sufficient deformation to the pores in the substrate, through heat, pressure or both, to allow the conducting particles to pass through them sufficiently to produce a conductive path.

The conductive material may be introduced by a tool which leaves an amount of the material on the surface on one or both sides which is moulded by the tool (not shown), or in a subsequent process, into a desired shape, for instance to form an electrical contact 28, either to further connection means intended to pass outside the cell or to another similar conductive assembly on a further substrate. The substrate 16 may have an electrode or connector track associated with it, preferably integral

with the substrate and formed on it be for example screen-printing or suction deposition.

In a preferred method, the substrate material is a porous fluoropolymer membrane, for example porous PTFE, and the impregnating material is polypropylene loaded with carbon particles. The melting point of the loaded polypropylene, less than 200°C, is significantly less than that of the PTFE (softening point around 300°C), allowing the polypropylene to be forced through the pores of the PTFE easily by a tool temperature of typically 200-240°C.

Example: For a PTFE sheet such as **Mupor** (Registered Trade Mark) type 131 (MUPOR Ltd., Alness, UK), thickness 0.2mm with mean pore diameter 2 μm, and impregnating material polypropylene loaded with 40 wt% carbon black particles of mean agglomerate dimension of order 200 nm (material from Whitaker Technical Plastics Ltd., Macclesfield, UK) good conductivity through the membrane was achieved using a hot pressing technique, at 200-240°C, and a pressure of approximately 200 N/cm^2, for 10s. This produced a low resistance contact through the membrane to a Pt/PTFE gas diffusion electrode, which was porous with mean pore size similar to that of the membrane, mounted on the opposite side of the membrane. Such an electrode and contact could be used in a gas sensor as shown in the embodiments described, to detect carbon monoxide.

Figure 3 shows a gas sensor 100 comprises a housing 102 with a reservoir 104 for liquid electrolyte. The reservoir 104 has at its upper end a support member 108 mounted on or attached to the housing to provide a rigid or semi-rigid support for the components connected thereto. The housing 102 has mounted in it contact pins 110, 114, 118 each in good electrical contact with associated moulded components of conducting polymer 112, 116, 120. Overlying the support member 108 is a first electrode assembly consisting of a membrane 122 with a catalyst layer 124. The catalyst material is preferably sintered together with the electrode to produce a robust electrode assembly. The catalyst layer is formed on the substrate prior to introducing the substrate into the housing, by for example screen-printing, suction

deposition etc. The catalyst layer 124 might be a porous layer formed from a catalytic material such as Pt or RuO2, bound together and to the substrate 122 by means of a PTFE binder as is known in the art. Alternatively it might be a nonporous material, for example a metal film, possibly treated to increase its catalytic activity. The substrate 122 is porous and is of a material of higher melting point than the material of the housing 102 and the conducting polymer 120.

The electrode assembly is sealed into the housing with catalyst layer 124 uppermost as shown, by for example application of heat and pressure, or ultrasonic welding. The housing material is locally melted and forced into the porous substrate 122 forming a strong bond in the regions 126. Simultaneously, the conducting polymer, which initially projects above the level of the housing surrounding it, melts and is forced through the substrate 122 and into contact with the electrode 124. If the catalyst is porous then the conducting polymer is preferably forced into the catalyst layer, so improving the electrical contact and physical robustness of the assembly.

A wick assembly 130 overlies the first electrode 124. The wick assembly is compressible and has extensions (shown as dotted outline 132) which reach down into the electrolyte reservoir. A second electrode assembly, consisting of one or more electrodes – two are shown in Figure 3, as 136 and 138 - on a second porous substrate 134, contacts the wick on the opposite side. At least the second electrode 136 consists of a porous catalytic layer capable of reacting signal gas in the presence of air and electrolyte.

The second electrode assembly is sealed to the housing with the catalyst layer lowermost, by application of heat and pressure, ultrasonic welding or similar means as before. The housing material is forced into the substrate 134 forming a bond in the regions 140 and the conducting polymer is melted and impregnated into the electrode 136 and any other electrode that is provided on the common substrate according to details of the embodiment, making electrical contact with them. This second process of sealing and making contact is essentially as described in the

Applicant's granted US patent US 5,914,019. Finally a housing cap 144 is mounted onto the housing 102, by heat sealing, ultrasonic welding or the like. Cap 144 provides access of gas from the exterior to the electrode 136 via the porous substrate 134 and a gas distribution space 148, that access being limited by a diffusion barrier 146, shown in the form of a capillary. The reservoir is partially filled with electrolyte (typically sulphuric acid) via a filling plug in the housing (not shown).

In use, second electrode 136 with gas access from the exterior, acts as the sensing electrode, and first electrode 124 acts as a reference electrode or in a two electrode cell, the counter electrode. If a third electrode 138 is provided, then this acts as the counter electrode.

Variations may be made, for example, the first substrate 122 might have two electrodes on it, with a second conducting polymer and pin contact arrangement to make contact to it, these electrodes functioning as the counter and the reference electrodes, and the second substrate 134 might have just one.

While the contact arrangements 114, 116 and 118, 120 are shown as being at different distances from the edge of the cell, these might be located in any practical geometry as suits the sealing process and tooling, for example, they might be in line with one another relative to the edge. Also, while the sealing surfaces 126 and 140 are shown as being at different levels in the cell, and the seal processes have been described as being done in two stages, especially if very thin components are used these surfaces might be at the same level, with compliance and flexibility of the components optionally being exploited to allow the seals to be made simultaneously.

The contacts are shown as being formed by a contact pin joined to the electrodes by a conductive polymer mass; alternatively, the pin might be absent and the

conductive polymer might itself lead to the outside of the cell, either with the conductive polymer co-moulded as part of the housing, or it may be bonded to the housing a separate components after moulding.

A further variation on this embodiment is in the design of the support means, shown as the support member 108. This could instead comprise a compliant component compressed between the base of the reservoir and the underside of membrane 122, with optional further sheet components to give even support to the components above it.

The stacked construction employed in the present invention reduces area used on the common substrate so reducing the "footprint" of the cell; secondly, in planar designs such as that in US 5,914,019 steps must be taken to prevent signal gas from reaching the reference electrode – this implies some form of seal between the edge of the gas distribution space 148 and the reference electrode. This seal is a source of unreliability and it is a great advantage to avoid need for it. The positioning of the reference electrode on the other side of a wick from the sensing electrode prevents signal gas from reaching it as (i) gas cannot diffuse quickly through the wick and (ii) most if not all signal gas will have reacted at the sensing electrode anyway.

Variation may be made to the aforementioned embodiments without departing from the scope of the invention. For example, for the sensors described herein, three or more electrodes may be formed on the membrane. These additional electrodes may generate a test gas so that the sensors have self-test capability.

Claims

1. A method of manufacturing a gas sensor having a housing containing a reservoir which in use receives an electrolyte, the method comprising the steps of: impregnating a gas porous membrane with a conductive material, so that said conductive material defines an electrical pathway between an electrical contact on a first surface of the membrane and an electrode on a second surface of the membrane and arranging the membrane to seal the reservoir.

2. A method according to claim 1 further including the step of attaching a wicking means (21) to the electrodes (14,16).

3. A method according to claim 2 whereby the wicking means (21) is pressed or sintered to the electrodes (14,16).

4. A method according to claim 3 whereby the wicking means (21) is sintered to the electrodes (14,16) at a temperature of between 300°C and 370°C.

5. A method according to claim 4 whereby the wicking means (21) is sintered to the electrodes (14,16) at a temperature of between 320°C and 370°C.

6. A method according to any preceding claim whereby the conductive material (26) is introduced into the substrate (18) via the wicking means (21).

7. A method according to any preceding claim whereby the conductive material (26) is introduced into the substrate (18) via an electrode (14,16).

8. A method according to any preceding claim whereby the conductive material (26) is introduced into the substrate (18) via the external connection means (28a,b).

9. A method according to any preceding claim whereby the conductive material (26) is introduced into the substrate (18) via the substrate (18).

10. A method according to any preceding claim whereby conductive material (26) in its melted state is introduced into the substrate (18).

11. A method according to any preceding claim whereby electrodes (14,16) and/or external connection means (28a,b) are formed on the substrate (18) by (a) screen printing, (b) filtering in selected areas from a suspension placed onto the substrate, (c) spray coating, or (d) sintering.

12. A method according to any preceding claim whereby the electrodes (14,16) are formed on the opposite faces of the substrate (18) to the external connection means (28a,b).

13. A method according to any of claims 1 to 8 whereby the electrodes (14,16) are formed on the same face of the substrate (18) as the external connection means (28a,b).

14. A method according to any preceding claim wherein the substrate (18) and the housing (12a,b) are bonded together using adhesive.

15. A method according to any of claims 1 to 10 wherein the substrate (18) and housing (12a,b) are bonded using heat and/or pressure so that material forming the housing melts and impregnates the substrate, thus forming a strong bond therebetween.

16. A method according to any preceding claim whereby the permeability of at least one region of the substrate (18) to gas is decreased in order to limit the amount of gas reaching an electrode.

17. A method according to claim 16 whereby the permeability of at least one region of the substrate (18) to gases is decreased by a) compressing the region, b)

impregnating the region(s) with wax, c) impregnating the region(s) with a polymer, or a combination of any of steps a) to c).

18. A gas sensor comprising: at least first and second electrodes formed on a planar substrate; a housing containing a reservoir which, in use, contains liquid electrolyte for contacting the first and second electrodes; an electrical contact for making external electrical connection from the gas sensor; and a conductive material disposed between an electrode and the external electrical contact, wherein at least a portion of the electrode and a portion of the substrate substantially adjacent thereto, is impregnated with the conductive material, the material forming an electrical pathway through the membrane which connects at least an electrode to the external electrical contact.

19. A gas sensor according to claim 18 wherein the electrodes (14,16) and/or external connection means (28a,b) are formed from a porous electrically conductive material containing catalyst material.

20. A gas sensor according to claims 18 and 19 wherein the first electrode (14) is a sensing electrode for creating the desired electrochemical reaction between the electrolyte (30) and the gas to be sensed.

21. A gas sensor according to any of claims 18 to 20 wherein the second electrode (16) is a counter electrode which performs an electrochemical reaction with oxygen.

22. A gas sensor according to any of claims 18 to 21 further including a reference electrode.

23. A gas sensor according to any of claims 18 to 22 further including a gas generating electrode.

24. A gas sensor according to any of claims 18 to 23 wherein the conductive mass (26) includes polymer electrolyte.

25. A gas sensor according to claim 24 wherein the conductive mass (26) is a plug, pin, or other shaped component suitable for forming an electrical path between the electrodes (14,16) and the external connection means (28a,b).

26. A gas sensor according to any of claims 18 to 25 wherein the external connection means (28a,b) includes polymer electrolyte.

27. A gas sensor according to any of claims 18 to 25 wherein the external connection means (28a,b) is a metal strip, or wire, which is attached to the substrate (18).

28. A gas sensor according to any of claims 18 to 27 further including a wicking means (21), the wicking means being arranged so that, in use, it contacts both the electrolyte (30) and the electrodes (14,16), thereby wetting the electrodes with electrolyte.

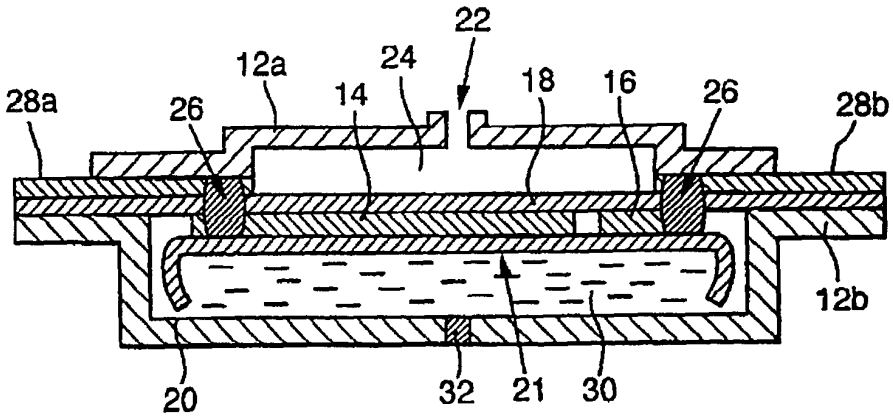
29. A gas sensor according to any of claims 18 to 28 wherein the wicking means (21) has at least one aperture formed therein through which polymer electrolyte (26) can be introduced.

30. A gas sensor substantially as described herein with reference to the accompanying drawing.

31. A method of forming an electrical pathway across a microporous membrane having first and second major surfaces; which membrane in use is impervious to liquid and permeable to gas, comprising the steps of: maintaining sufficient heat to melt a conductive material; urging the melted conductive material through pores of the membrane at a first surface by establishing a pressure differential across the surfaces; controlling the heat and pressure differential until the conductive material emerges at the second surface; and allowing the material to cool so as to form a continuous, electrically conductive pathway from the first to the second surface whilst preserving the liquid impermeability and gas permeability characteristics of the membrane.

10a

Fig.1.



10b

Fig.2.

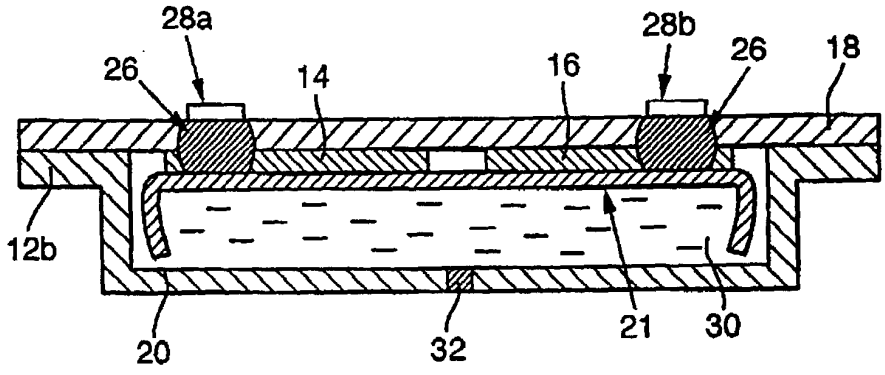
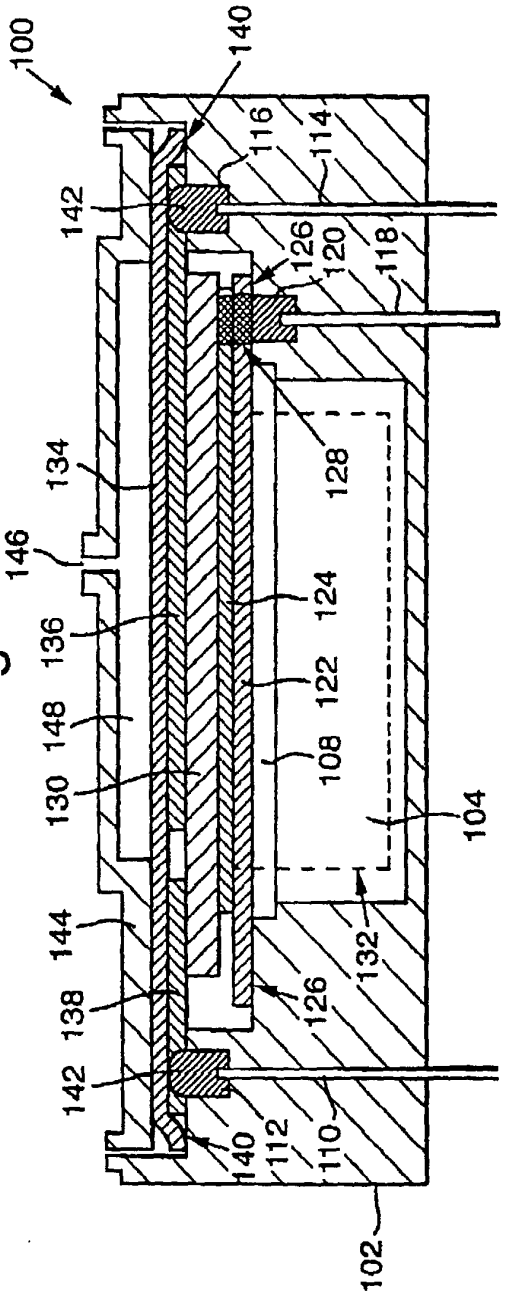


Fig.3.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